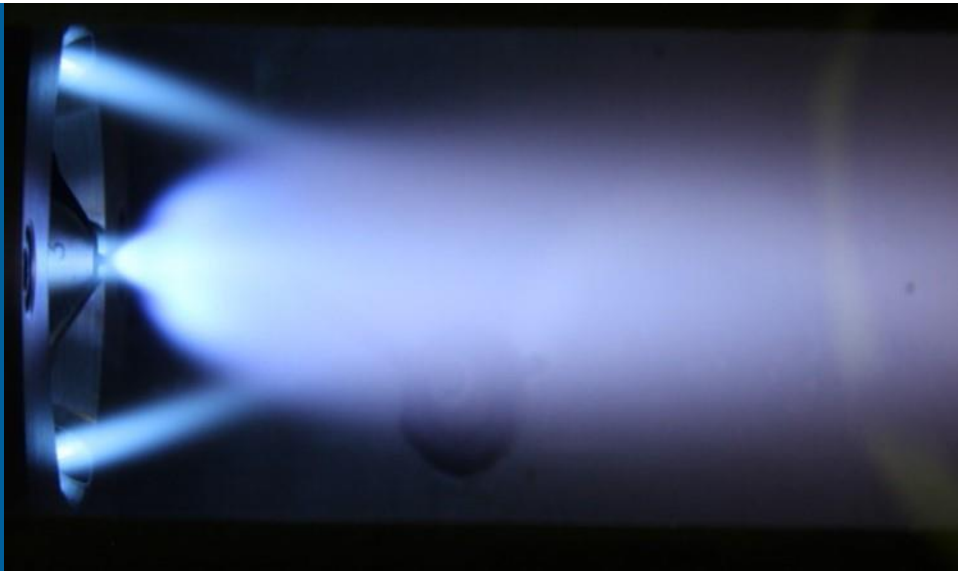
PROCESS R&D FOR DROPLET-PRODUCED POWDERED MATERIALS



JOSEPH LIBERA (PI)

Project ID: BAT315

2022 DOE Vehicle Technologies Office Virtual Annual Merit Review (AMR),
June 1-4, 2022,

Overview

Timeline
- Project Start Date: September 2016
- Project End Date: September 2022

Budget
- Total project funding:
  - $400K in FY21

Barriers
- **Cost of high-energy Li-ion**
- **Life**

Partners
- Cabot Corporation
- ORNL, LBNL
- Purdue University

Supporting battery research for:
- DOE Battery Research Community

Objectives - Relevance

- To develop aerosol processes to produce cathode active materials, solid electrolytes, additive particles for life extension and filler particles for polymer composites.

- The relevance of this task to the DOE Vehicle Technologies Program is:
  - This synthesis technique has the potential to provide large cost reduction through continuous high-volume production methods.
  - The high purity and crystallinity of FSP materials has the potential to improve performance for the same materials synthesized by other means.

Approach and Strategy

- Flame Spray Pyrolysis is a proven industrial technology for commodity scale production of numerous simple compounds (TiO_2, C black, SiO_2). The ANL FSP facility provides a highly instrumented pre-pilot powder production facility for the development and optimization of aerosol production of powders. This heavily instrumented facility provides in-operando scientific feedback to enable rapid materials development and fundamental understanding of this complex manufacturing process.

- Complimentary aerosol techniques: (a) Spray Pyrolysis, (b) Slurry Flame Spray Pyrolysis, (b) Slurry Spray Pyrolysis, and (c) Dry Aerosol Calcination

- Maintain a close relationship with our industrial partners to assure we follow sensible routes for potential commercialization.

Approach - Milestones

FY21	Design and build scale up for 500 g/hour FSP production	*Ongoing*	July-2022
	Single Crystal Cathode development	*Ongoing*	Sept 2022
	Dry Aerosol Calcination	*Complete*	Jan 2022
	Feasibility study for LLZO green powder based SSB architectures	*Ongoing*	Sept 2022

Technical Accomplishments And Progress Overview - Summary

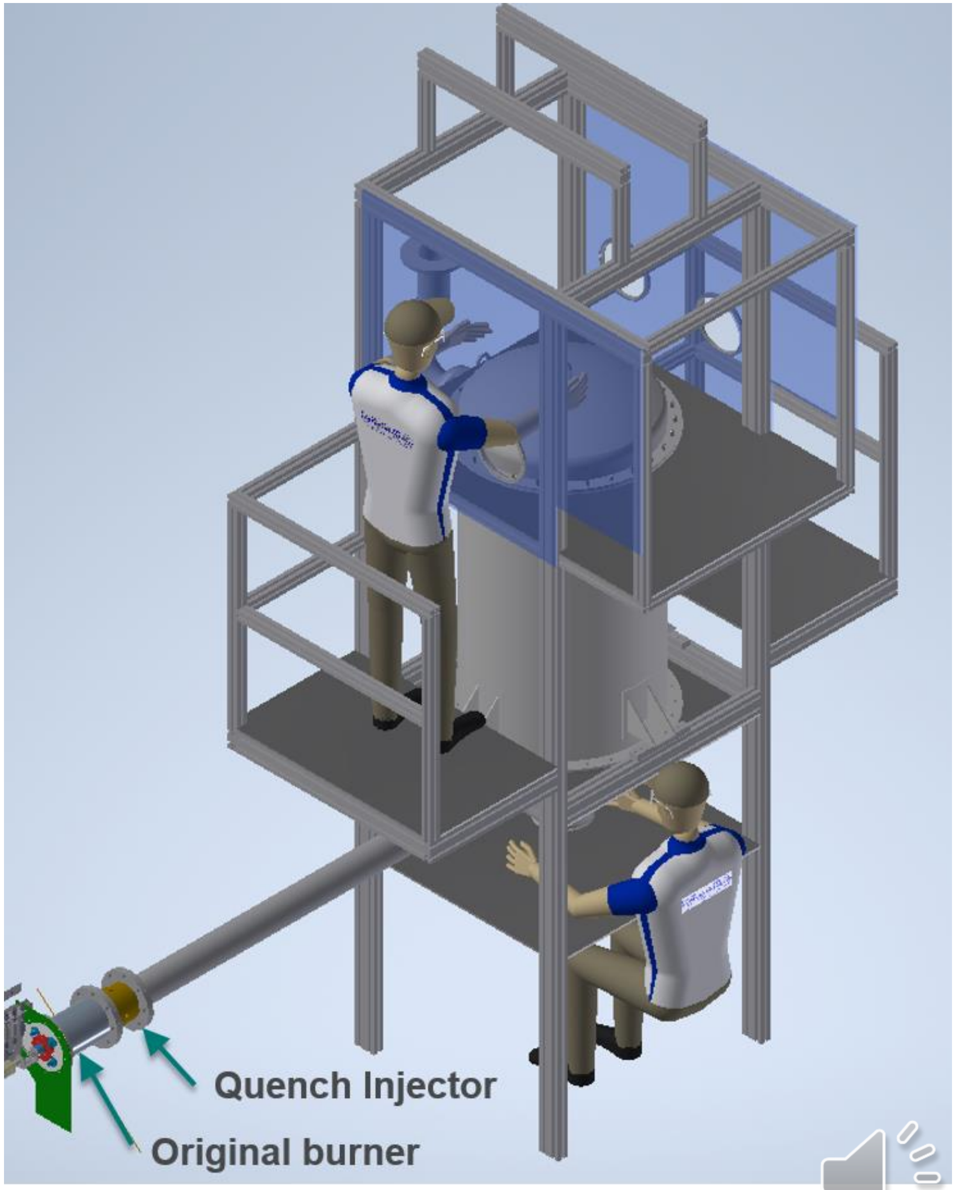
- Completed Design and Major Procurements for 500 g/hour upscale of the FSP synthesis facility.

- Added dry aerosol processing capability and tested dry aerosol calcination of spray pyrolysis green powders

- Optimized calcination protocol for layered phase formation of high Ni spray pyrolysis green powders.

Technical Accomplishments And Progress Overview – FSP Facility Scaleup

- **Design for 500 g/hour facility completed.**
- **Procurement of baghouse completed and received in May 2022.**
- **500 g/hour production for LLZO**
- **250 g/hour production for additive nanoparticles such and NCM 111, alumina**
- **Back-pulsing baghouse for continuous operation.**
- **Engineered controls for nanopowder handling.**
- **Positive pressure combustion to eliminate blower suction requirement for collection.**
- **Uses the same liquid feed, combustion gas feed and burner as original 50 g/hour system.**
- **Close-coupled gas quenching system.**
- **No tube furnace due to size and space restrictions**

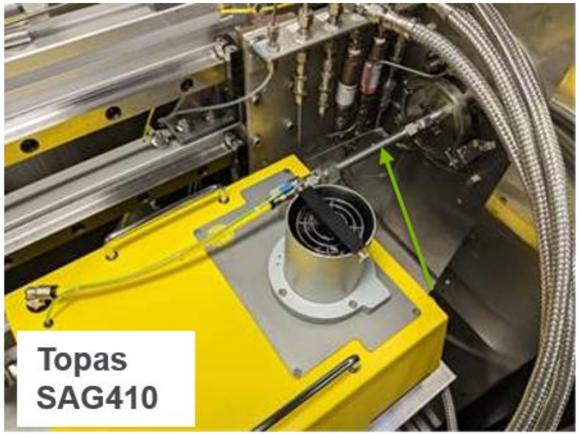


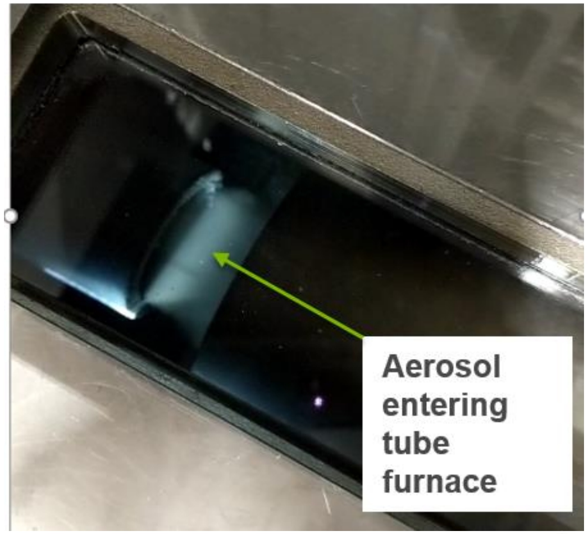
Technical Accomplishments And Progress Overview – Dry Aerosol Calcination Capability

Air → SAG 410 → Tube Furnace → Collection
Dry Powder → SAG 410

- Dry Aerosol Calcination concept:
  - Contactless sintering – preserves particle morphology during sintering
  - Rapid sintering – optimized gas exchange per particle
  - Reduces porosity and surface area of individual particles or clusters.

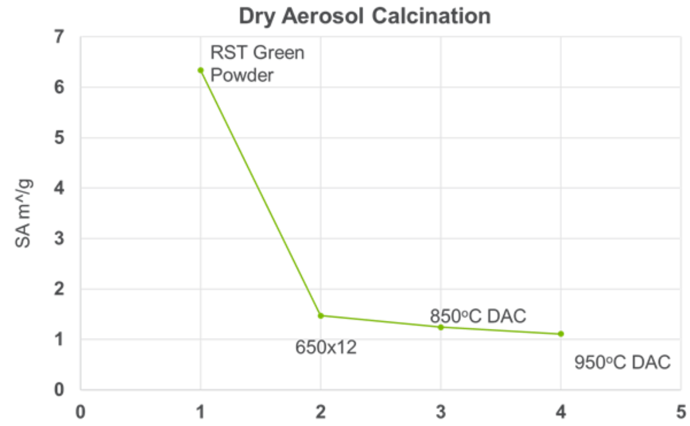
- Aerosol loading is 2-325 g/m^3

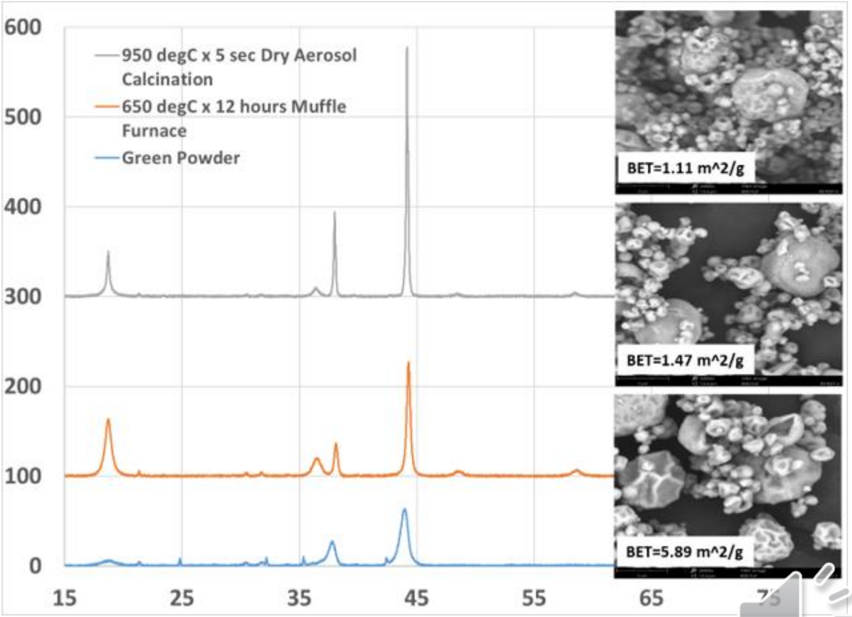




Technical Accomplishments And Progress Overview – Dry Aerosol Calcination Capability

- Dry Aerosol Calcination was tested on Spray Pyrolysis NCA green powder
- The result was compared to a 12 hour static calcination in a muffle furnace
- The residence time of the DAC aerosol in the tube furnace is <5 sec.
- With the tube furnace at 950ºC the dry aerosol shows an similar outcome as the 12-hour static calcination.
- Densification and surface area reduction was demonstrated while preserving the original particle morphology and avoiding sintering
- Layered phase not achieved. May require 2nd pass with inter-stage gas exchange to remove CO_2 and water from 1st pass.





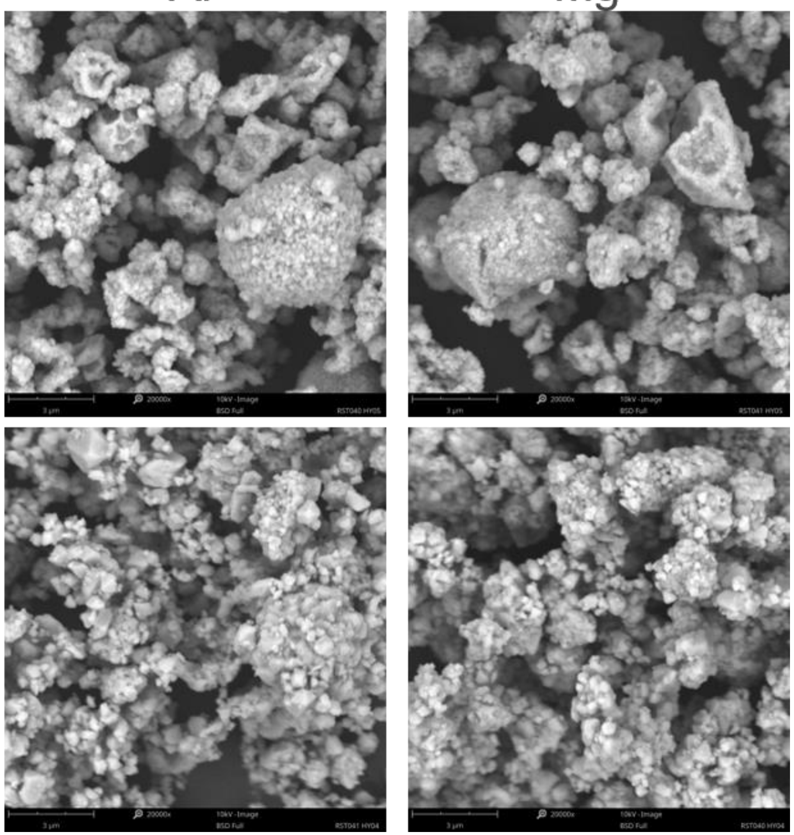
Technical Accomplishments And Progress Overview - LNO by Spray Pyrolysis Particle Morphology

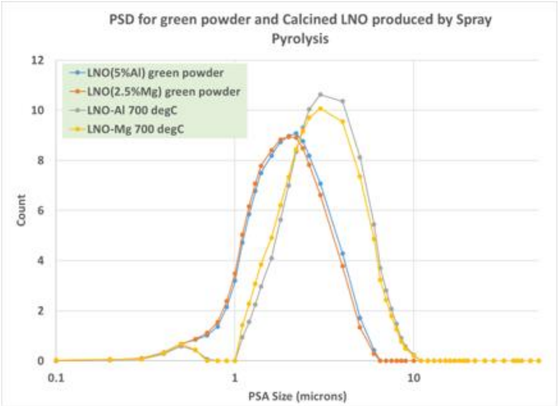
- Green powders were synthesized using Spray Pyrolysis of nitrate solutions of Li, Ni and Al or Mg.
- Calcination of Al- and Mg-doped Lithium Nickel Oxide was optimized
- 700ºC vs 730ºC reveals a threshold to crystallinity and brittleness. 730ºC calcination results in substantially more fines after de-agglomeration processing (mortar milling). Mg doping results in much higher brittleness than Al doping.

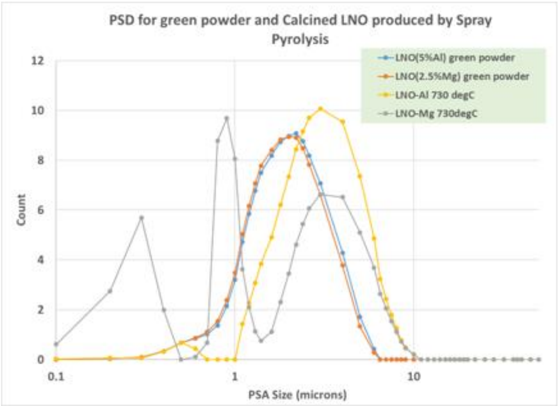
Al Mg

700ºC

730ºC







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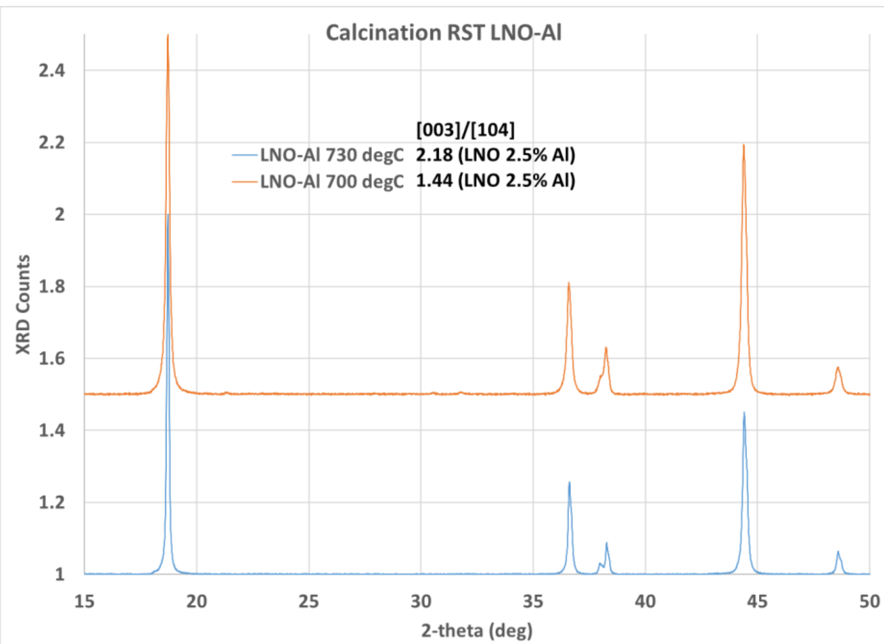
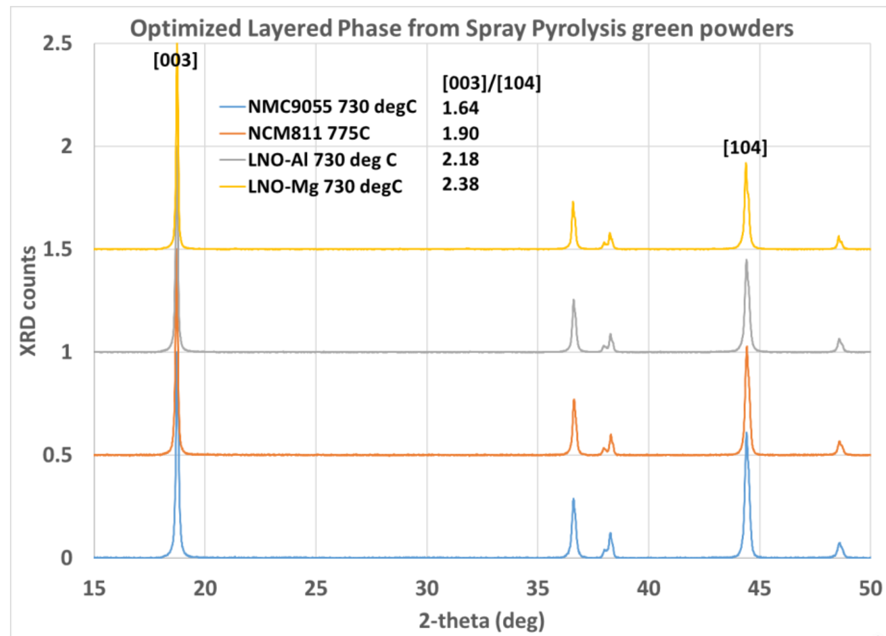
Technical Accomplishments And Progress Overview - LNO by Spray Pyrolysis Calcination Properties

- 700ºC vs 730ºC reveals lesser crystallinity and slight impurity. Despite this, the 700ºC samples exhibit better capacity retention compared to 730ºC.
- Higher Ni samples in aerosol generated green powders require lower temperature to achieve good layered phase.





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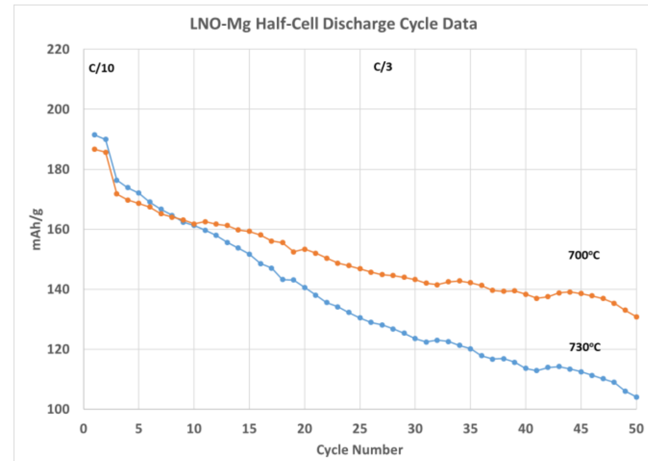
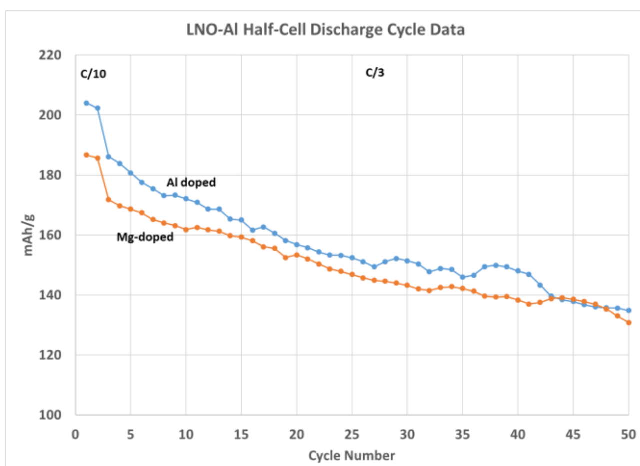
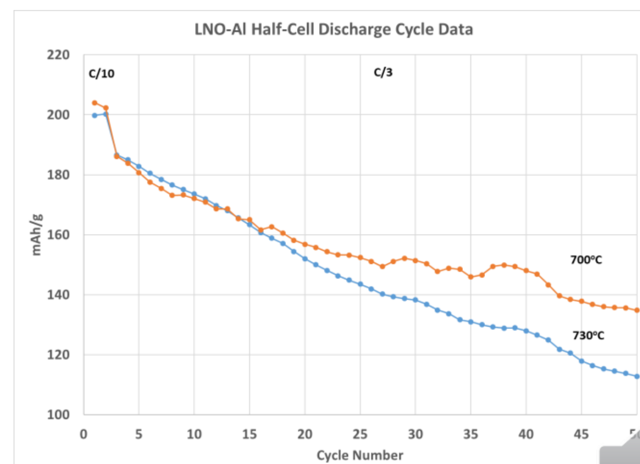
Technical Accomplishments And Progress Overview - LNO by Spray Pyrolysis Battery Performance

- Half-cell data for cycling between 2.7-4.3V with 6.8 mg/cm^2 loading
- 700ºC vs 730ºC reveals lower capacity fade for both Al- or Mg-doped LNO, possibly due to more fines in the 730ºC calcined samples.
- Mg-doped LNO has lower starting capacity than Al-doped LNO but exhibits a higher capacity retention.







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Collaboration and Coordination with Other Institutions

- Cabot Corp. is a continuing partner in low-Co cathode active phase development.

- ANL completed sponsorship a CRI Innovator Volexion who graduated to startup status. Volexion develops novel graphene-active material composite cathode architectures.

- Collaboration with Purdue University for the development of polymer/particle composite electrolyte.

Remaining Challenges and Barriers

- Optimization of aerosol processes for cost-competitiveness.

Proposed Future Research

- Develop additive particles for extension of battery life

- Develop filler particles for polymer composite electrolyte applications

- Develop dry aerosol processing for reduced cost cathode thermal processing
  - Develop coating add-ons to allow for life-extension over-coating of cathode powders

- Provide production of LLZO with 500 g/hour FSP unit.

Summary Slide

- Completed design and major purchases for 500 g/hour FSP scale up unit.
- Installed and commissioned dry aerosol processing capability and demonstrated operation for cathode thermal processing.
- Demonstrated Al- and Mg-doped LNO synthesis using Spray Pyrolysis.

Acknowledgement

- The PI gratefully acknowledges Peter Faguy and Dave Howell for their continued support of aerosol processing research for energy materials